SOLUBILITY OF NOBLE GASES IN LIQUID ALKALI METALS[1]

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ABSTRACT

It has been carried out the review of experimental data availablein literature on solubility of noble gases in liquid alkali metals andtheir alloys as per in effect on 2000. It should be noted that this re-view offers a clear illustration of the fact that the solubility of helium, argon and xenon in melted sodium and potassium is basically investigated in it.

As regards to other liquid alkali metal - noble gas systems they have been either insufficiently fully investigated or havenot been studied at all. Besides the temperature and pressure interval measurements to be carried out is not considerably broad and is bounded by 1000 K temperature and 1 Mpa pressure. Proceeding from this analysis the conclusion can be drawn on the necessity of a wide spectrum of experimental investigation to carry out on solubility of noble gases in melted alkali metals in broad ranges of temperature and pressure. In view of these factors as such measurements are not realized so far theoretical calculation of solubility of noble gases in liquid alkali metals are recommended to carry out at temperatures up to 2000 K and pressures up to 10 Mpa.

KEY WORDS: Genry’s coefficient; experimental data; melted alkali metals; noble gases; solubility.

1. INTRODUCTION

The nuclear power which has received a large development, has beenput forward at present in the first place the problems of reliabilityand safety of atomic engineering. It needs to be recognized that thatin its turn induced the necessity to carry out experimental investigation of different heat-hydraulic processes in atomic reactors, their stability and an effect of great number of phisicochemical factors.

Thus, both the presence of shielding noble gas in the first circuit gaseous spaces of the fast neutron nuclear reactors and the emergence in its path of other noble gases which are reaction products of nuclear fuel, leads to dissolution of these gases in the liquid-metal heat-transfer agent. The changing of the reactor load leads to the change of the heat-transfer agent temperature, and as a results of it the noble gases dissolved in the heat-transfer agent can be flushed out of a metal melt and exists in its volume in the form of bubbles of one or another size. The latter circumstance undoubtedly has produced a considerable effect on the heat exchange conditions and the flow hydrodynamics in the fast neutron nuclear reactor fuel element.

It is worth mentioning that if some metals are really exposed to radiation by neutrons, then in this case many of such nuclear reactions lead to the formation of noble gases [1]. It is reasonable to assume that the following nuclear reactions at light elements is of great practical interest:

$$n + {}^{6}Li \rightarrow {}^{4}He + {}^{3}H \qquad n + {}^{9}Be \rightarrow 2n + {}^{8}Be$$

$$\downarrow \qquad\qquad\qquad\qquad \downarrow$$

$$^{3}He; \qquad\qquad\qquad\qquad {}^{4}_{2}He + 94 keV;$$

$$n + {}^{9}Be \rightarrow {}^{4}He + {}^{6}He \qquad\qquad n + {}^{10}B \rightarrow {}^{7}Li + {}^{4}He.$$

$$\downarrow$$

$$^{6}Li;$$

The formation of helium in these reactions, as well as xenon ^{139}Xe and krypton ^{85}Kr isotopes as the reaction products of fast neutron reactor nuclear fuel, has raised a question to study the solubility heat-transfer agents of these reactors [2].

It can be asserted that the same problem has occured in thermonuclear fusion as well [3]. The coalescence reaction of deuterium and tritium

$$^{2}D + {}^{3}T \rightarrow {}^{4}He(3.52 MeV) + n(14.06 MeV),$$

which is the most close to a practical realization, leads to the formation of high energy neutron, and it combines with the material of the first thermonuclear reactor wall - lithium. Helium and hydrogen isotopes have been formed as a results of such interaction.

It should be noted that the study of noble gas - melted metal system is of great theoretical interest. This can be explained by the monoatomic nature of noble gases and the lack of chemical activity in the ratio to melted metals that makes it possible to study their disso-
lution process, ignoring chemical reactions. Viewed in this light, noble gas atoms can be considered as solid uncharged spheres, and their properties are analogous to those vacancies in a solid body. There is a good reason to believe that such an approach has given an opportunity to use the temperature dependence of noble gas solubility in metal melts for detection of structure changes.

2. EXPERIMENTAL DATA ON SOLUBILITY OF NOBLE GASES IN LIQUID ALKALI METALS

The main information on experimental works, available in the literature up to 2000, which were devoted to the investigation of noble gas solubility in liquid alkali metals, are listed in Tables Nos.1 and 2.

Henry’s coefficient* temperature dependences for helium - liquid alkali metal and noble gas - sodium melt system to be some literature data generalizations, are shown in Fig.1 and 2. Moreover Henry’s coefficient values at these plots are presented in a logarithmic scale. Since as shown in Fig.1 and 2 the solubility of noble gases in melted alkali metals is increased with the temperature (and pressure) growth. Perhaps it should be stressed that its value at both constant temperature and pressure is the more the alkali metal atomic weight, and the less noble gas atomic weight.

The following conclusions may be drawn, proceedings from the analysis of experimental investigations of noble gases solubility in liquid alkali metals:

1. The number of investigation noble gas - alkali metal melt systems is not great. It was basically measured the solubility of helium and argon in liquid lithium, sodium, and potassium. As regards to other systems they have been insufficiently studied or have not been studied at all.

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* The solution of noble gases in liquid alkali metals can regarded as infinitly diluted one, as concentration of the solute (gas) is small in comparision with concentration of the solvent (liquid alkali metal). For these solutions to be at a temperature T and pressure P, Henry's law is acting to interconnect between f_2 volatility (and at small pressure - between P_2 partial pressure as well) of the dissolved gas and its x_2 molar concentration in solution: $f_2(P,T)=K(P,T)\,x_2$.

Coefficient K is called Henry's coefficient, as it present solution property, which depends on the temperature and pressure. In practice Henry's coefficient K_H is usually used as equal to inverse coefficient K, i.e. $K_H(P,T)=1/K(P,T)$.

2. The solubility values of noble gases in melted alkali metals are small, and this naturally makes it difficult to carry out experimental investigations. That is why, the error of measurements to be made constitutes no less than 20-25% and even more. It is interesting to note that the disparity between the data obtained by different researches for the same system turns out to teach even the same order of magnitude.

3. The investigation parameter range bounded by 1000 K temperature and 1 MPa pressure however does not meet practical requirements nowadays.

3. CONCLUSION

It should be recognized that at noble gas - liquid alkali metalsystem is not fully investigation, and the measurements to be made clearly indicate that the temperatures and pressures are not relatively high, as well as a high error of their results, and this is not in conformity with modern high standarts of science and engineering. In view of these factors new high-precision broad-based experiments are required to carry out at temperatures up to 1500-2000 K and pressures up to 10 MPa.

At the same time under these circumstances it is required to develop the theory of noble gas solubility in alkali metal melts on the basis of the analysis of available in the literature theories on gas solubility in liquids. The value of solubility calculated with the help of such a theory at high temperatures and pressures would make it possible to compare theoretical an experimental results.

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Table 1. Experimental Data on the Solubility of Noble Gases in Liquid Alkali Metals

Author(s),	Gas-metal	Range of parameters of investigation		Purity	
publication year(s), reference(s)	studied system	T, K	P, MPa	gas, % vol.	metal, % mass
C.R. Mitra, 1956-1961, [4, 5]	Xe-Na	392-412	1 atm	-	-
R.E. Cleary et al., 1962-1965, [6 - 8]	He-Li He-K	922-1144 755-977	0.12-0.31 0.12-0.31	- -	- -
S.K. Dhar, 1963-1964, [9-12]	Ar-Na Kr-Na	613-753 478-753	0.30-0.94 0.12	- -	- -
H.W. Savage et al., 1965, [13]	Ar-(Na+K)	1005	0.17	-	-
E. Veleckis et al., 1967-1976, [14-26]	He-Na Ar-Na Xe-Na	623-823 602-804 623-873	0.20-0.88 0.14-0.69 0.25-0.82	99.999 99.999 99.999	- - -
K. Thormeier, 1969-1970, [27-30]	He-Na Ar-Na	573-873 623-873	0.10-0.39 0.05-0.39	- -	99.98 99.98
M.N. Arnoldov et al., 1970, [31,32]	He-(Na+K)	573	1 atm	99.981	99.77
R.A. Blomquist et al., 1971-1976,	Kr-Na	673-823	0.71	-	-

[33,34]					
N.I. Bets et al., 1972, [35]	Ar-Na	460-673	0.11-0.20	-	-
A.G. Mozgovoy et. al., 1998, [36]	Ar-Na	1273	1	99.999	99.99

Table 2. State of Experimental Studies of Solubility of Noble Gases in Liquid Alkali Metals

Alkali metal	Noble gas					
	He	Ne	Ar	Kr	Xe	Rn
Li	[6-8]	-	-	-	-	-
Na	[16-20,24,25, 27-30]	-	[9-12,14,15,24, 25,27-30,35]	[33,34]	[4,5,21-23, 26]	-
K	[6-8]	-	-	-	-	-
Rb	-	-	-	-	-	-
Cs	-	-	-	-	-	-
Fr	-	-	-	-	-	-

FIGURE CAPTIONS

Fig.1. Temperature dependence of Genry's coefficient for helium-melted alkali metal system: 1 - lithium according to experimental data [6-8]; 2 - sodium [16-20,24,25]; 3 - sodium [27-30]; 4 - potassium [6-8].

Fig.2. Temperature dependence of Genry's coefficient for noble gas - liquid sodium system: 1 - helium according to experimental data [14-20,24,25]; 2 - helium [27-30]; 3 - argon [27-30]; 4 - argon [14,15,24,25]; 5 - krypton [33,34]; 6 - xenon [21-23,26].

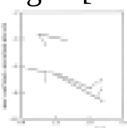


Fig. 1

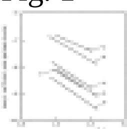
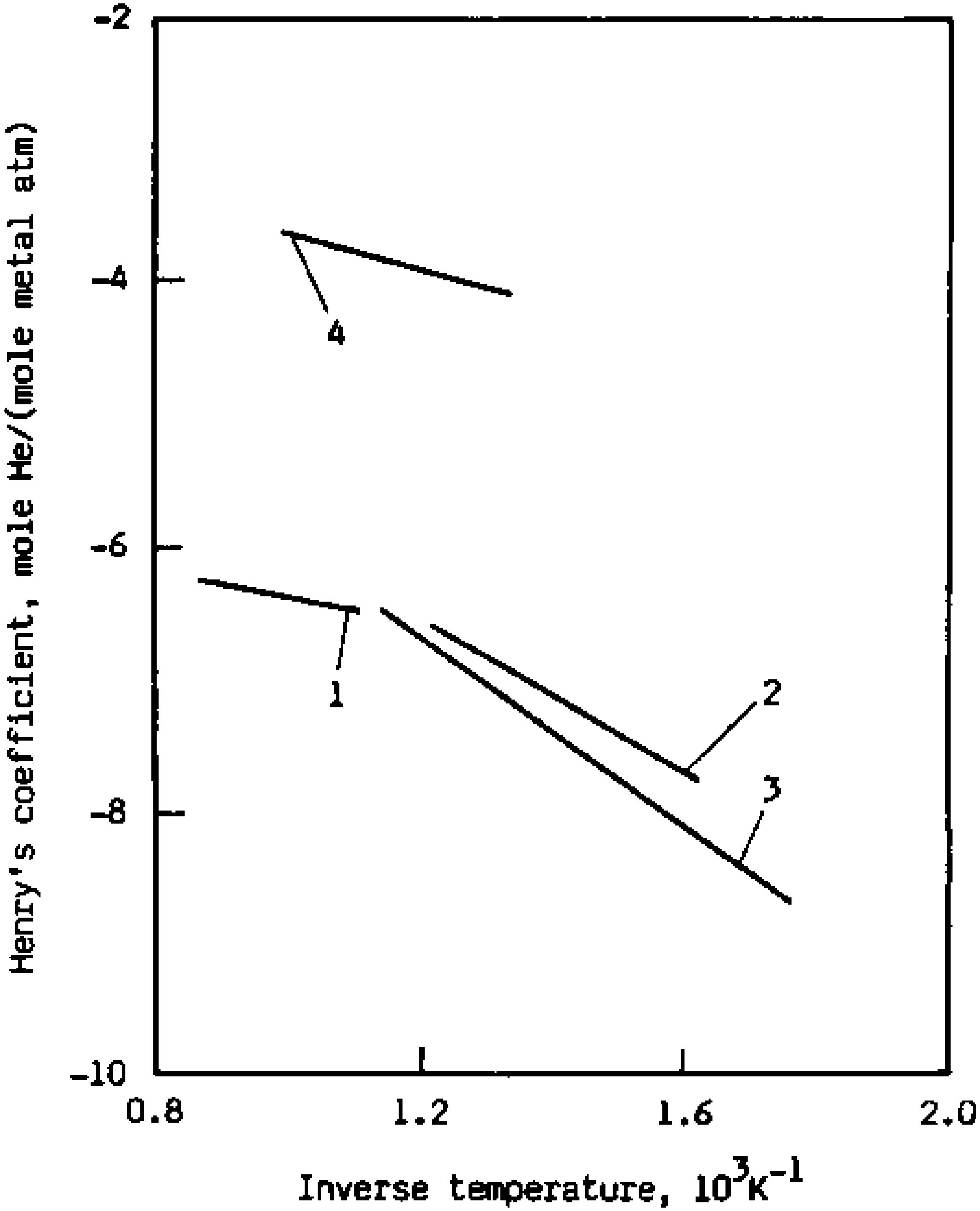


Fig. 2.



-2
-4
-6
-8
-10
0.8
1.2
1.6
2.0
Henry's coefficient, mole He/(mole metal atm)
Inverse temperature, $10^3 K^{-1}$
1
2
3
4

Fig.1

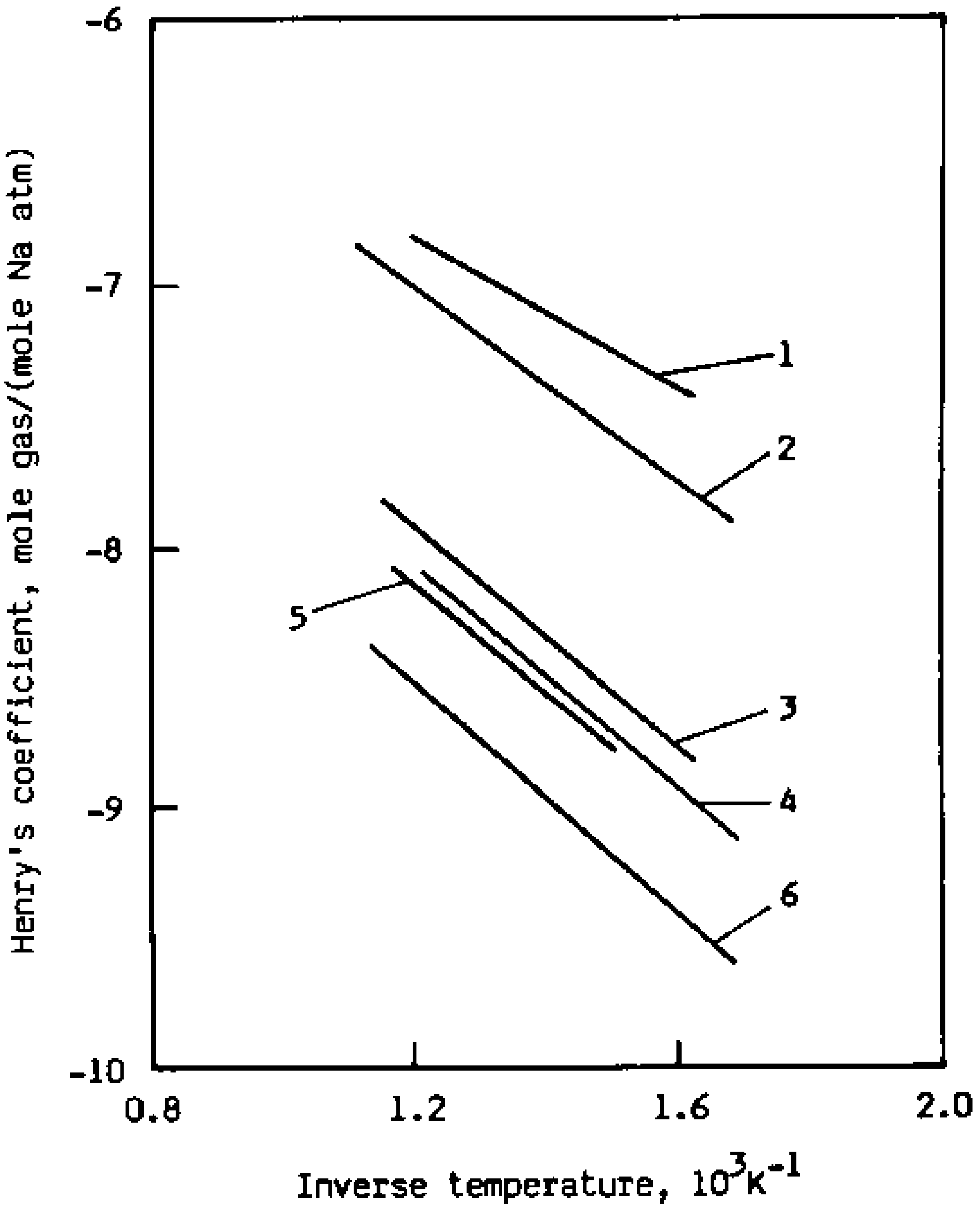

-6
-7
-8
-9
-10
0.8
1.2
1.6
2.0
Henry's coefficient, mole gas/(mole Na atm)
Inverse temperature, 10^3 K^-1
1
2
3
4
5
6

Fig.2